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## (54) IMPROVEMENTS RELATING TO PLASTICS SHEET MATERIAL

- (71) We, PASTONA (JOHN WADDINGTON) LIMITED, a British Company, of Wakefield Road, Leeds LS10 3TP, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—
- This invention relates to plastics sheet material, and in particular concerns the production of a plastics sheet material which has a wide range of uses, because of the properties thereof.
- As is well known, plastics sheeting is now in extensive use in many industries, but heretofore there has not been provided a plastics sheet material which has general application in a large number of fields. Generally speaking, particular plastics materials are designed for particular uses, and materials which are designed for a particular use, usually have limited other uses.
- The material which is provided by this invention will have, we feel, a very wide range of uses. For example, we have found that it is extremely suitable as a substitute for cardboard, and paper in general, and yet it can also be used to form thin walled containers such as drinking cups or foodstuffs tubs and lids therefor.
- In endeavouring to provide a plastics sheet material which has a wide range of uses, we have concerned ourselves with introducing additives to achieve a suitable material which can be formed into sheet for example by extruding and/or calendering and we believe that by the present invention we have provided a material of a unique and novel composition which forms into sheet form satisfactorily, and the resulting sheet has a wide range of applications.
- In accordance with the present invention there is provided a method of manufacturing sheet material comprising compounding 5-70 parts by weight of inorganic particulate material with 95-30 parts by weight of polypropylene; polypropylene-ethylene sequential copolymer; (as herein defined) polypropylene-ethylene random copolymer; high density polyethylene; or any mixture of these, the compounding being carried out essentially by mechanical working in a mixing apparatus to generate heat, and to disperse the inorganic material evenly throughout the resin so as to produce a molten mass of uniform consistency having a melt flow index in the range 0.55 and 2.2 cc/10 minutes inclusive at 230°C under a load of 2.16 Kg passing the molten mass directly to an extruder apparatus, and extruding the mass either directly into the sheet in a thickness of 0.1-1.2 mm or to a calender apparatus which calenders the mass into the sheet in a thickness of 0.1-1.2 mm.
- The mechanical working of the inorganic material and the resin is an important part of the present invention as it is necessary that the inorganic material should be very evenly dispersed throughout the resin as otherwise it will not form into satisfactory sheets. The heat to produce the molten mass comes essentially from this working although where the working is effected by means for example of a screw working in a barrel, the barrel may be encased in electric heating bands which may supply some heat to the mixture, but essentially are provided to prevent the heat generated by the mechanical working from escaping from the barrel and by passing the molten mass directly to the extruding apparatus. There is minimum heat loss and minimum heating requirements during the heating stage.
- Conventional mixing apparatus may be used, such as contra-rotating and intermeshing screws for compounding the resin and inorganic material.
- The resin polypropylene blended with compatible elastomer in such quantity as to confer upon the sheet material similar impact existence and flexibility characteristics as are possessed by sheets produced from polypropylene-ethylene sequential copolymer (as herein defined)

According to a preferred arrangement of the invention, the resin is

- (a) a sequential copolymer (as herein defined) of propylene with from 10 to 35% by weight (based on the weight of the copolymer) of ethylene and/or
  - (b) a blend of polypropylene having sufficient of a rubber to confer on the blend an impact resistance and flexibility which are within the range of impact resistances and flexibilities as are possessed by the range of sequential copolymers defined in (a).
- 15 Folded containers and other folded articles and blanks produced from the material made by this preferred feature of the invention possess most of the desirable properties of high quality cardboard and that an average shopper would find great difficulty in realising the containers are not made from cardboard. Further, the sheet material from which the containers are made is inherently water-proof and so does not need a waterproofing treatment. The sheet material will also lie flat in the container and blank making machinery. These characteristics are obtained without the need specifically to subject the sheet material to an orientation treatment prior to creasing and folding. The sheet material of this invention preferably has a thickness of at least 0.1 mm if the folded containers are to have properties comparable to those of high quality cardboard and it is preferred to use sheet material having a thickness of at least 0.4 mm which scores and cuts neatly enough to enable it to compete with high quality cardboard in high speed packaging machinery.
- 40 This sheet material can also be thermally formed satisfactorily into a whole host of small domestic tubs, and cups and lids therefor. The sheet material is preferably extruded from a composition comprising a sequential copolymer of propylene and ethylene and/or a blend of polypropylene and rubber as stated herein. By "sequential copolymer" we mean a particular type of copolymer which is made by first polymerising propylene and then prior to the completion of the polymerisation reaction, ethylene is injected into the polymerisation zone so that as the polymerisation proceeds, polymer chains are produced which become increasingly rich in copolymerised ethylene randomly distributed among the polymerised propylene. These sequential copolymers in sheet form are softer and more flexible than polypropylene yet when mixed with particulate inorganic material and used for sheets, the sheets develop an impact resistance and a rigidity which are adequate in comparison with high quality cardboard, and at the same

time the sheets are capable of enough orientation to permit neat creasing and folding and also thermoforming. Moreover, these properties are obtained with sequential copolymers which are high melt flow index materials and are not therefore high molecular weight materials.

Polypropylene (i.e. propylene homopolymer) can be used to make sheets having impact resistance and flexibility similar to sheets made from the sequential copolymers described above, by blending with a rubber. Examples of rubbers which may be used are polyisobutylenes, butyl rubbers and ethylene-propylene elastomers such as those described on pages 255 to 258 of "Chemistry and Industry" of 16 March 1974. The precise quantities of rubber needed to confer the required impact resistance and flexibility on the polypropylene sheets will vary from rubber to rubber and can be determined by routine impact and flexibility tests.

There should preferably be 20 to 70% by weight of the solid particulate inorganic material and such inorganic material desirably has a hardness of less than 5.5 on the Moh's scale. Examples of suitable inorganic materials may include talc, calcium carbonate, dolomite, kaolin or gypsum or any combination of these. Use of an inorganic material which gives the sheet a soft feel is desirable to facilitate creasing and hinging both of which appear to cause a compression in the sheet material. Such inorganic material also appears to have a desirable effect on the feel of the sheet material. The introduction of soaps and oils into the composition from which the sheet material is formed may well improve the production of the material. The preferred inorganic material is talc or calcium carbonate; good results have been obtained using both the Chinese talc known as Haichen talc and calcium carbonate.

The particles of the inorganic material preferably should be capable of passing through ASTM Sieve 140 and preferably 97% by weight of the particles should be capable of passing through ASTM Sieve 325. The preferred materials should preferably comprise at least 30% by weight of particles having a largest dimension of between 10 and 18 microns in order to promote good creasing and hinging.

The composition from which the sheet material is formed may also comprise from 1 to 8% by weight of a pigment having a hardness of less than 6.8 on Moh's scale.

The presence of the pigment achieves a uniform background on which to print. The preferred pigment is titanium dioxide. Anatase titanium dioxide has a hardness of 5.5 to 6 on Moh's scale and has been used to good effect. However, rutile titanium

dioxide, which has a hardness of 6 to 6.5 causes less long term degradation of the sheet material and may be preferable if the sheet material is to be used in making hinged containers or other articles which are expected to have a long life.

Both the rutile and anatase titanium dioxide pigments should preferably be coated with up to 5% by weight of alumina and up to 2% by weight of silica based upon the weight of the pigment. The pigment may be used in combination with up to 0.5% by weight of an optical brightener such as ultramarine.

The composition from which the sheet material is formed, preferably by extrusion but also by calendering may optionally contain processing aids such as soaps including calcium stearate. The compositions may also contain conventional additives such as stabilisers.

The surface of the sheet materials used in the performance of this invention has a good ability to receive printing inks. However, this ability can be further enhanced by subjecting the surface to one of the oxidation treatments of the type described in the book "polythene" edited by Renfrew and Morgan and published by Iliffe, see pages 542 and 543 of the 2nd edition.

The most convenient of these treatments is the one which uses a corona discharge.

The composition preferably includes one or more antioxidants and it is suggested that conventional amount of hindered phenol thio-ester be used.

A particular embodiment of the invention is illustrated by the following example:—

A sequential propylene-ethylene copolymer comprising 15% by weight of copolymerised ethylene was made by polymerising propylene and then injecting 15% by weight of ethylene into the polymerisation zone before all of the propylene had polymerised. Conditions were chosen so as to produce a copolymer which had a melt flow index of 1.1 cc/10 minutes.

A thermoformable composition was produced by mechanically mixing together in a compounding machine, 55.9% by weight of the sequential copolymer 40% by weight of a particulate talc, 4% by weight of a titanium dioxide and 0.1% by weight of calcium stearate, so as to cause thorough dispersion of the inorganic material throughout the resin and to heat the mixture into a molten mass of even consistency. The talc was a Haichen talc and 98% by weight of the particles of the talc were capable of passing through ASTM Sieve 325 and 33% by weight of the particles had a maximum dimension within the range 10 to 18 microns. The titanium dioxide was

an anatase titanium dioxide comprising 1.5% by weight (based on the  $\text{TiO}_2$ ) of alumina and 0.7 by weight of silica in the form of a surface coating on the particles of titanium dioxide.

The composition obtained from the compounding machine was passed directly to an extruder from which it was extruded into a sheet material having a thickness of 0.8 mm. It was found that the sheet material was capable of lying flat and was useable in the form of cut and creased blanks in high speed machinery used to make folded cardboard containers from blanks. Another feature of the material was that it could be cut and creased on conventional cutting and creasing machinery without the use of a make ready, thereby facilitating the setting up of the machinery, and reducing the cost of processing the material. The sheet material compared well with high quality cardboard in folding, creasing, scoring, perforating and cutting operations and could be folded to form a container having a crush resistance similar to that of a high quality cardboard container. The sheet material also had good dead-fold properties and could be easily glued. The folded containers made from the sheet material had a pleasant feel and were very receptive to printing ink with the result that to a casual observer they were almost indistinguishable from folded containers made from high quality cardboard.

Moreover, the material thermoformed extremely well into domestic cups and tubs and lids therefor, such as are used for the storage of food products such as margarine, butter, jam and so on.

Furthermore, it has been found that the material of the invention satisfactorily receives markings by ballpoint pens, solvent based markers, pencils, type-writing and printing, the inorganic material giving the sheet material a certain amount of surface absorbency. In order to enhance the receptiveness of the material to such markings, it may be subjected to flame treatment or corona discharge treatment.

It is believed that as an alternative to talcum and/or chalk in powdered form any one of a mixture of the following may be used:—clay; calcium carbonate; stearate (coated if desired) kaolin; calcium silicate; asbestos; barytes; gypsum; mica.

In the manufacture of the sheet of the invention, this is done in a continuous process starting from the raw materials, namely the resin and inorganic particulate material, the one or more anti-oxidants, and any other additives, required for the purposes of the end use of the sheetings, and compounding the raw materials, and then leading the molten mass directly while

soft, to an extrusion apparatus or calender apparatus where it is formed into sheet.

The extrusion die is attached directly to the compounding apparatus so that a composite apparatus performs the two functions, namely compounding and extrusion into sheet.

The melt flow index of the molten mass is determined according to the proceeding of British Standard 2782: Part 1/105C/1970 on standard melt flow index equipment, which normally measures in weight the material which flows out of an orifice time period at certain conditions of temperature and weight applied, but because the introduction of the inorganic material into the resin so greatly affects the specific gravity of the mass as compared to neat resin, it is better for this invention to procure results by determining the volume of material per time period. Furthermore, the volume which flows through the orifice is a direct indication of the viscosity of the mass, and it has been found that only material of viscosity in the range indicated by the melt flow index of 0.55 cc-3-2.2 cc/10 minutes inclusive at 230°C under a weight of 2.16 Kg form satisfactorily into sheet material having a wide range of uses.

Owing to the special selection of the melt flow index in accordance with the present invention the material has excellent flexing and handling characteristics, and furthermore it is possible using conventional cutting and creasing machinery, to produce carton blanks which are extremely satisfactory as explained above, the material creasing along the crease lines in a manner almost identical to conventional cardboard.

The sheet material readily receives embossing and retains such embossing in permanent set.

The sheet material according to the invention which is cut and creased to form carton blanks can be erected into cartons by conventional equipment, and the blanks can be provided with locks as are conventional cardboard blanks, or hot melt adhesive can be used to hold the cartons in erected condition. Conventional board or paper glueing equipment can be used for the application of hot melt adhesives.

The material therefore can be used effectively in all cases where carton blanks have been provided, without any, or at least, any substantial, modification of the existing cardboard erecting and glueing equipment.

Another suitable application for the sheet material is for the production of playing cards, and for the production of such articles it is desirable to ensure that the opacity of the material is made as high as possible.

The material satisfactorily receives var-

nishing using normal varnishing techniques such as are used for the fixing of printing inks on sheet material or for the fixing of other impressions on such material.

The sheet material can be thermoformed satisfactorily and can be used for the production of thin walled containers and lids therefor which are produced by a thermoforming or equivalent process. In such process the sheet material will require to be heated. This may be effected conventionally, but preferably will be in accordance with any one of our co-pending patent applications 07647/74 and 21174/74 (Serial No. 1,475,145). The material is sufficiently stable, and the use thereof results in a high quality products of good stability.

The sheet material is unaffected by most water and solvent based liquids and the material furthermore presents a reasonably high barrier to moisture vapour.

This makes the material extremely good for containers which have to hold hygroscopic or water containing products.

If desired, the material may be laminated with another material or coated to vary the surface finish thereof.

It is to be appreciated that the amount of filler in the plastics material may be varied as desired, within the range specified, and the material may also include as explained herein and in varying degrees, other additives which may be desirable for the particular end use of the material.

The sheet material of thickness at the higher end of the range can be used to produce jackets for binders, and other more rigid articles, especially articles in which it is desirable to provide a hinge. The examples given in this specification are in no way intended to limit the use of the material.

It has been found that the material according to the invention extrudes in an extremely satisfactory manner and is of high quality. Furthermore, where the material is used for producing articles which are cut from the material, leaving a skeletal waste, this waste can be re-used and can be returned, suitably comminuted, to the compounding apparatus.

In a particularly interesting modification of the invention blowing agents as well as inorganic particulate material are incorporated into the sheet material. The blowing agents are incorporated in most cases as a minor amount such as 0.4% by weight of the molten mass of a solid chemical substance capable of decomposing into gases at a temperature somewhat less than the temperature of sheet formation. Of these chemical substances a typical substance is p-toluene sulfonyl semicarbazide.

The effect of the blowing agent is to cause the specific gravity of the resultant

extruded sheet to be considerably decreased, in other words the sheet as expanded.

Reference will now be made to the accompanying drawing, of which Fig. 1 shows diagrammatically how compounded material is produced and Fig. 2 shows diagrammatically how container blanks are produced from the sheet material as described in the example above.

Referring to Fig. 1 a compounding machine of conventional form is indicated by reference numeral 1, and it will be seen that this is associated with feed hoppers 2 and 3. The resin, namely the sequential copolymer resin is introduced into the compounding machine at hopper 2. Whilst the inorganic particulate material is introduced at hopper 3. In the compounding machine, the materials, including any additives such as antioxidants, colouring, matter and oils are mechanically worked into a molten mass of melt flow index in the range as herein specified and the molten mass is passed directly to an extruding machine 10 and is extruded in sheet form 12 (shown in Fig. 2).

Referring now to Fig. 2, the extruding machine is indicated diagrammatically again by reference numeral 10, and the sheet material being extruded therefrom by reference numeral 12. A break is shown in the sheet material 12 between the extruding machine 10, and a conventional cutting and creasing machine 14, in order to indicate that the sheet material may well be stored for example in reel form before being passed to the cutting and creasing machine 14. Indeed, the extruding may be done in one location or factory, and the cutting and creasing done in another location or factory. It is on the other hand possible to pass the sheet material 12 directly from the extruder to the cutting and creasing machine 14, assuming that the material has cooled and set sufficiently to enable cutting and creasing to be carried out thereon. Furthermore, it may be more usual to cut the material 12 into large sheets which are individually cut and creased as is done conventionally with high quality cardboard.

In any event, the material 12 is shown emerging from the machine 14 as having been cut and creased to define container blanks 16, the skeletal waste of the sheet material being indicated by numeral 18. This skeletal waste can be re-processed by being ground to particle form and returned to the compounding machine in order to maximise use of the material.

In the example shown, each blank comprises a number of rectangular panels connected together by crease lines formed in the machinery 14, and one of the panels is provided with end closure panels 20, another

of the panels being provided with a glue tab 22. The container which can be erected from each of the blanks 16 is also shown in the figure, and the erection is by a conventional process by gluing the tab 22 to the outside of the extreme panel at the other side of the blank. To close the container, the end panels 20 are simply folded over as indicated by the arrows in the figures and tuck in tabs of these end panels serve to hold the end panels in closed position.

It will be appreciated that the machinery 14 can be adapted to produce more complicated blanks, or even simpler blanks such as might provide book jackets or wallets which do not require any glueing, or the blanks may simply define inserts for insertion in other packaging containers.

This invention also provides a method for making folded containers, other articles or blanks therefor, according to this invention using machinery conventionally used in the manufacture of folded cardboard containers other articles or blanks therefor, with or without a make ready.

It is also possible, as shown in the figures to produce thermoformed containers, such as tubs and cups as indicated at 24 in the drawing, from the sheet 12. Conventional thermoforming method may be used.

Several more specific plant installations will now be described.

1. Polypropylene powder complete with stabilisers etc., and filler, for example the chalk or talc, referred to herein, are taken from bulk storage silos and automatically fed to a high speed mixer.

The thoroughly mixed ingredients are then fed directly into a twin contra rotating screw extruder where the molten polymer and filler are subject to high shear action, and the resulting homogeneous compound forced through a standard flat sheet die to produce the sheet material. The extrudate is then passed through a conventional sheet line consisting of a polishing roll stack, surface treatment unit, such as a Corona discharge unit, anti-static bath, haul off and slitter, terminating in a winding unit for reels or a guillotine and stacking unit for flat sheets.

2. One alternative is to take the polypropylene powder with stabilisers etc., and feed this with the chalk or talc filler directly into a compounding unit, such as a Buss Ko Kneader. The Buss Ko Kneader would then be fitted with a conventional cross head extruder, this cross head extruder being fitted with a standard sheet die and conventional sheet line equipment as described in (1).

The maximum temperature in the compounding and extrusion systems is in the order of 250°C.

A Buss Ko Kneader type P.R. 200 and extruder arrangement is capable of producing up to 1,000 kgs per hour of 40% filled polypropylene in sheet form.

5 An 80 mm dia. twin screw extruder is capable of producing up to 250 kgs per hour of 40% filled polypropylene in sheet form.

10 This application is divided out of cognate application No. 19469/75 and 24533/75 (Serial No. 1,554,143) which relates to sheet material, and said cognate application claims sheet material of which examples are described herein. Additionally, application No. 942/78 (Serial No. 1,554,147) and Application 946/78 (Serial No. 1,554,149) are divided out of the said cognate application. Application No. 942/78 (Serial No. 1,554,147) discloses an expanded sheet material made from a mass which includes a blowing agent, and an example of such sheet material is described herein. Application No. 946/78 (Serial No. 1,554,149) relates to a foldable sheet material article, of which examples are described herein.

#### WHAT WE CLAIM IS:—

30 1. A method of manufacturing sheet material comprising compounding 5-70 parts by weight of inorganic particulate material with 95 to 30 parts by weight of polypropylene; polypropylene-ethylene sequential copolymer (as herein defined);  
35 polypropylene-ethylene random copolymer; high density polyethylene; or any mixture of these, the compounding being carried out essentially by mechanical working in a mixing apparatus to generate heat, and to disperse the inorganic material evenly throughout the resin so as to produce a molten mass of uniform consistency having a melt flow index in the range 0.55 and 2.2 cc/10 minutes inclusive at 230°C under a load of 2.16 Kg passing the molten mass directly to an extruder apparatus, and extruding the mass either directly into the sheet in a thickness of 0.1-1.2 mm or to a calender apparatus which calenders the mass into the sheet in a thickness of 0.1-1.2 mm.

55 2. A method according to Claim 1 wherein the resin is polypropylene blended with compatible elastomer in such quantity as to confer upon the sheet material similar impact resistance and flexibility characteristics as are possessed by sheets produced from polypropylene-ethylene sequential copolymer (as herein defined).

60 3. A method according to Claim 1 or 2, wherein the resin is (a) a sequential copolymer (as herein defined) of propylene with from 10 to 35% by weight (based on the weight of the copolymer) of ethylene

and/or (b) a blend of polypropylene having sufficient of a rubber to confer on the blend an impact resistance and flexibility which are within the range of impact resistances and flexibilities as are possessed by the range of sequential copolymers defined in (a). 70

4. A method according to Claim 3, wherein the resin is said blend of polypropylene and the rubber is polyisobutylene butyl rubber and an ethylene-propylene elastomer. 75

5. A method according to any one of the preceding claims, wherein the solid particulate inorganic material has a hardness of less than 5.5 on the Moh's scale. 80

6. A method according to claim 5, wherein the solid particulate inorganic material is capable of passing through sieve 140 as defined in ASTM Designation E11-61. 85

7. A method according to claim 3, wherein 97% by weight of the particles of inorganic material are capable of passing through ASTM Sieve 325. 90

8. A method according to claim 5, 6 or 7 wherein at least 30% by weight of the particles of inorganic material have a largest dimension in the range 10 to 18 microns inclusive. 95

9. A method according to any preceding claim, wherein the inorganic material is free from fibrous components.

10. A method according to any preceding claim, wherein the sheet material includes soap to reduce the co-efficient of friction of the particles of inorganic material one relative to another. 100

11. A method according to claim 10, wherein the soap includes calcium stearate. 105

12. A method according to any preceding claim, wherein the inorganic particulate material includes one or more of the following; talc, calcium carbonate, dolomite; kaolin and gypsum. 110

13. A method according to any preceding claim, wherein the sheet material includes from 1 to 8% by weight of a pigment having a hardness of less than 6.8 on Moh's scale. 115

14. A method according to claim 13, wherein the pigment is titanium dioxide.

15. A method according to claim 14, wherein the titanium dioxide is anatase titanium dioxide. 120

16. A method according to claim 14, wherein the titanium dioxide is rutile titanium dioxide.

17. A method according to claim 14, 15 or 16, wherein the titanium dioxide is coated with up to 5% by weight of alumina and up to 2% by weight of silica based upon the weight of the titanium dioxide. 125

18. A method according to any claims 13 to 17, wherein the pigment is used in 130

combination with up to 0.5% by weight of optical brightener.

19. A method according to any one of the preceding claims, wherein the surface  
5 of the sheet material is subjected to a known oxidation treatment.

20. A method according to claim 19, wherein surface of the sheet material is subjected to a corona discharge.

10 21. A method according to any one of the preceding claims wherein the composition includes the anti-oxidant hindered phenol thioester.

15 22. A method according to any preceding claim, including a minor amount of a blowing agent.

23. A method according to Claim 22 wherein the blowing agent is introduced in solid form with the compounding operation in producing the molten mass.

20 24. A method according to claim 22, or 23, wherein the blowing agent is introduced in an amount equal to 0.4% by weight of the molten mass.

25 25. A method according to claim 24, wherein the blowing agents P-toluene and sulfonyl semicarbazide.

26. A method according to any preceding claim, wherein the sheet material is cut and printed to define playing cards.

27. A method according to any of  
30 claims 1 to 25, wherein the sheet is formed into articles by a heating and shaping process.

28. According to any one of claims 1 to 25 wherein the sheet is cut and creased  
35 to form packaging container or other article blanks.

29. A method according to claim 28, comprising cutting and creasing the sheet material using conventional cutting and  
40 creasing machines.

30. A method according to claim 29, wherein cutting and creasing is performed without the use of a make-ready.

31. A method according to claim 1 and  
45 substantially as hereinbefore described.

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